Assessing Performance of GCR Shielding Materials for Deep Space Missions

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Acknowledgements to Co-workers

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Performance of Shielding Materials in Terms of Dose to Spacecraft Crew

- Liquid H_2
- Liquid CH_4
-
- Polyethylene (CH_2)
-
- H_2O
-
-
- Al—Inadequate shielding
-
-
- Pb
-

Best

Worst

Potential range for new and multi-functional shielding materials: polymer-infiltrated carbon foams and fiber bodies; polymer composites; CH_4 adsorption on carbon materials; (hydrides and hydride/carbon or hydride/polymer composites)

Approaches for Improved Shielding via Materials Science

- Increase H content and decrease higher Z content--some improvement relative to polyethylene is available but possibilities are limited
- Fabricate high-H material to serve multiple functions--e.g., manage heat transfer, bear structural loads, serve as debris shields, store fuels and fluids,…These materials could possibly replace materials in current use, simultaneously reducing biological dose as well as spacecraft mass

Integrate Materials Science and Particle Transport Assessments of Shielding Materials

- **Materials experiments**
  - Carbon foams infiltrated with polyethylene
  - Carbon fiber monoliths infiltrated with polyethylene
  - Polymer fibers and composites
  - Methane adsorption on carbon
- **Shielding performance**
  - Irradiations at NSRL with 1 and 0.6 GeV/nucleon O^{16}
  - Analysis of beam fragmentation
  - GCR shielding calculations for all experimental materials and benchmarks

Materials

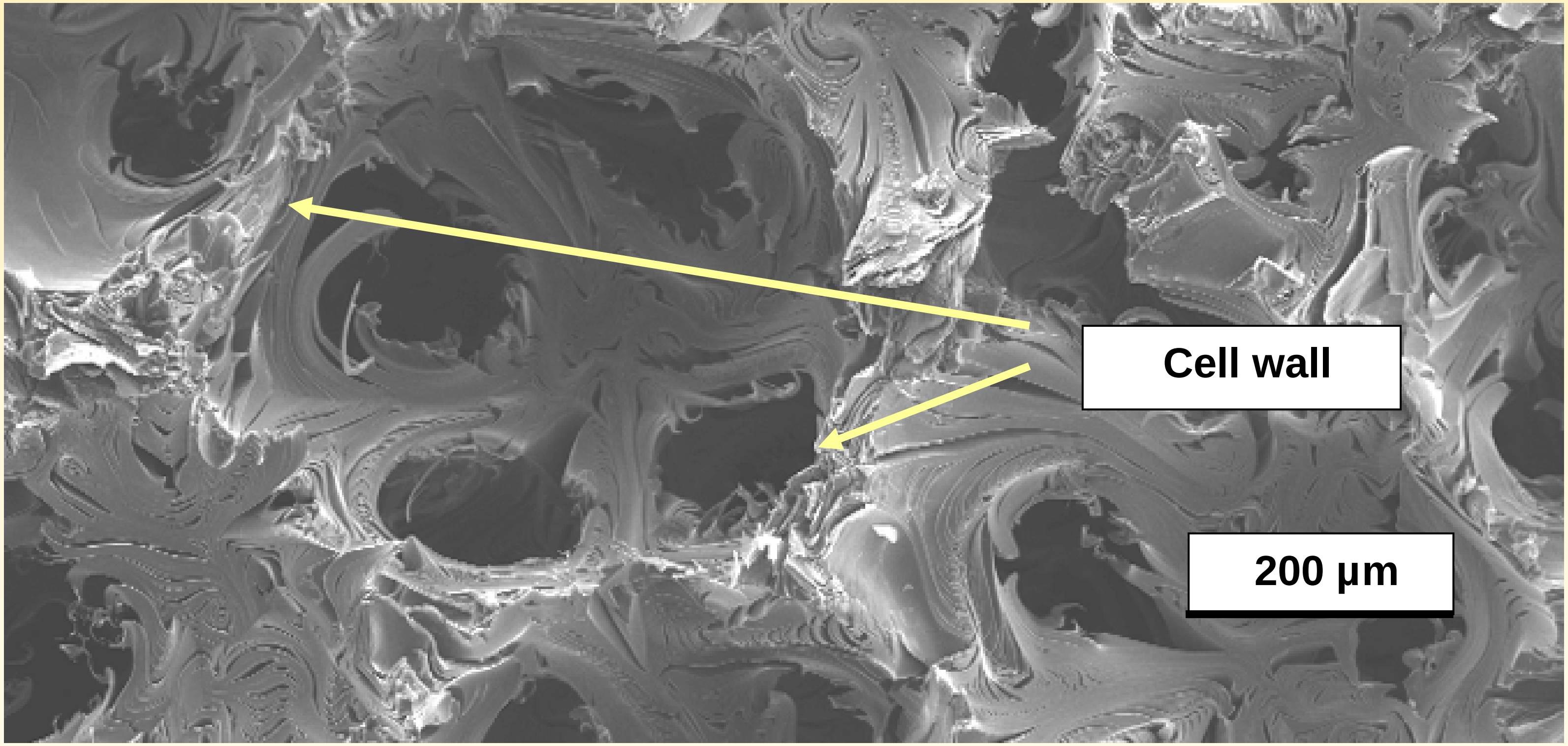
- **Investigate shielding properties of two classes of materials**
  - Carbon forms infiltrated with PE
  - Polymers and polymer composites
- **Both classes of materials have potential to serve one or more functions in addition to GCR shielding**
- **Carry out fabrication of the materials**
- **Prepare specimens as multiple disks or layers, allowing thicknesses exposed to beam to be varied according to the irradiation plan**

Carbon Materials before PE Infiltration

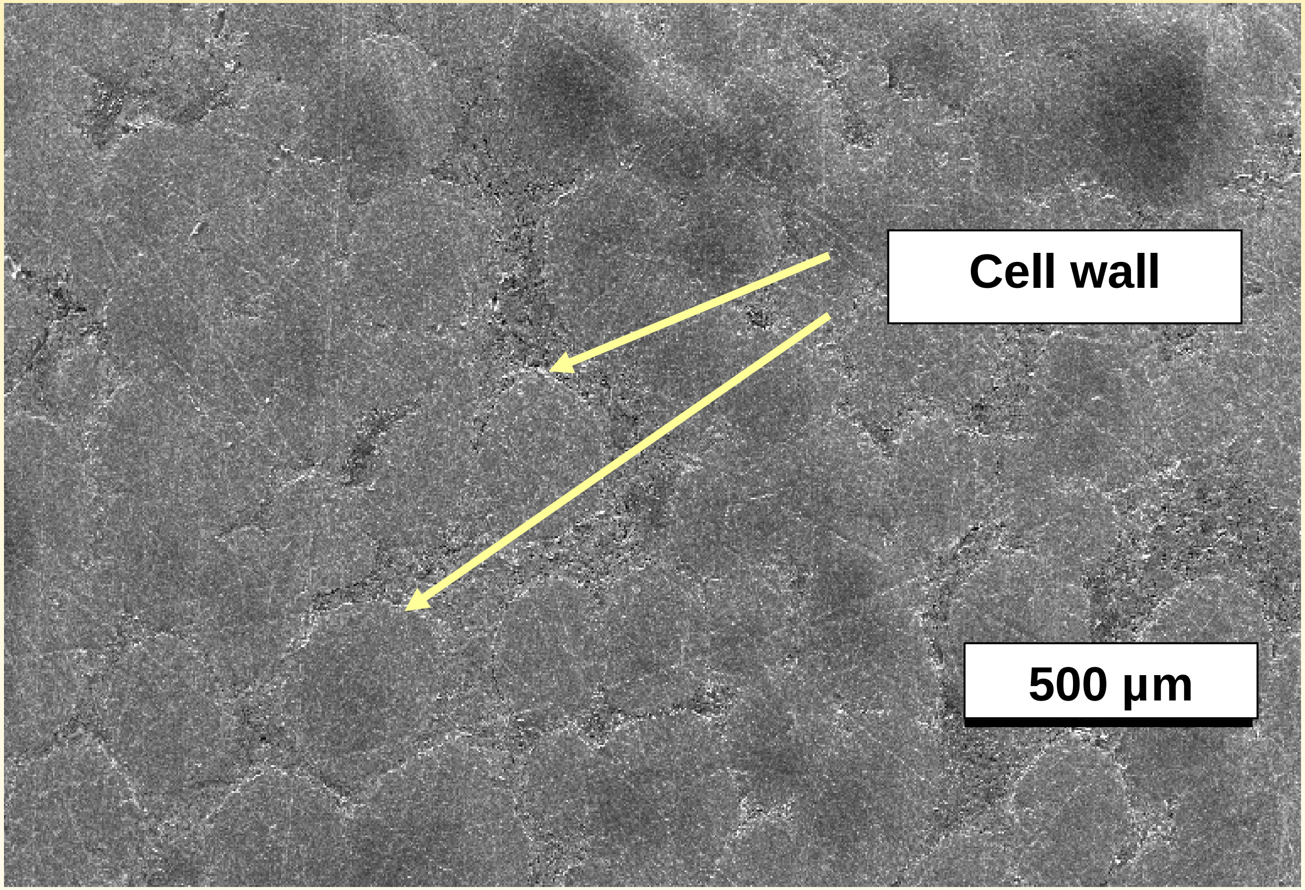
Material Description*	Density (g/cm^3)	Void Vol. (%)	K ($W/m{\cdot}K$)	σ_c (MPa)
Lo Dens. Foam (LF)	0.25	86.8	50	0.8
Hi Dens. Foam (HF)	0.52	72.6	100	2.0
Lo Dens. Fiber Monolith (F)	0.35	81.6	0.14-0.30	1.7-2.0

*Abbreviations LF, HF, and F appear in subsequent plots.

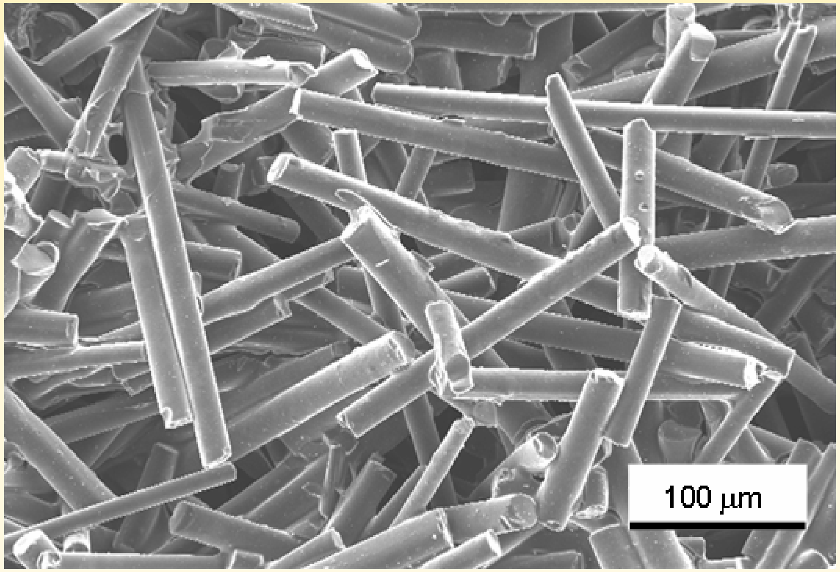
Low Density Carbon (Graphitic) Foam Prior to Infiltration

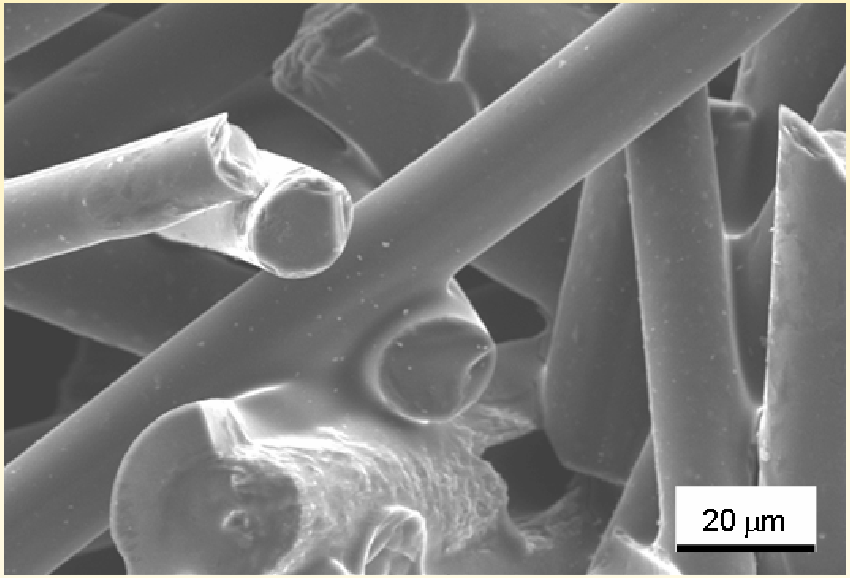


Low Density Carbon (Graphitic) Foam After Infiltration with Polyethylene



Carbon Fiber Monolith before Infiltration with Polyethylene

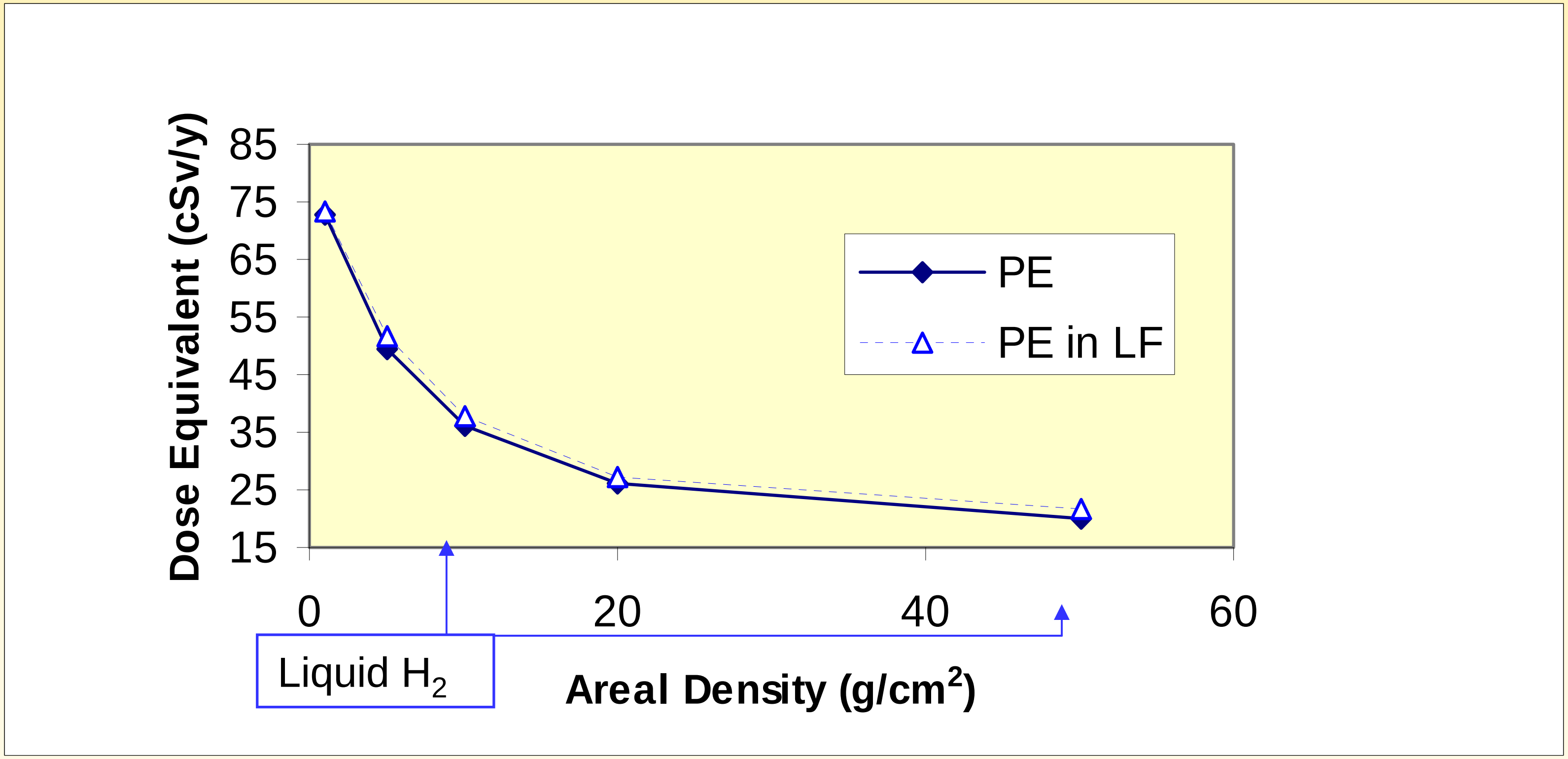




Materials Compositions

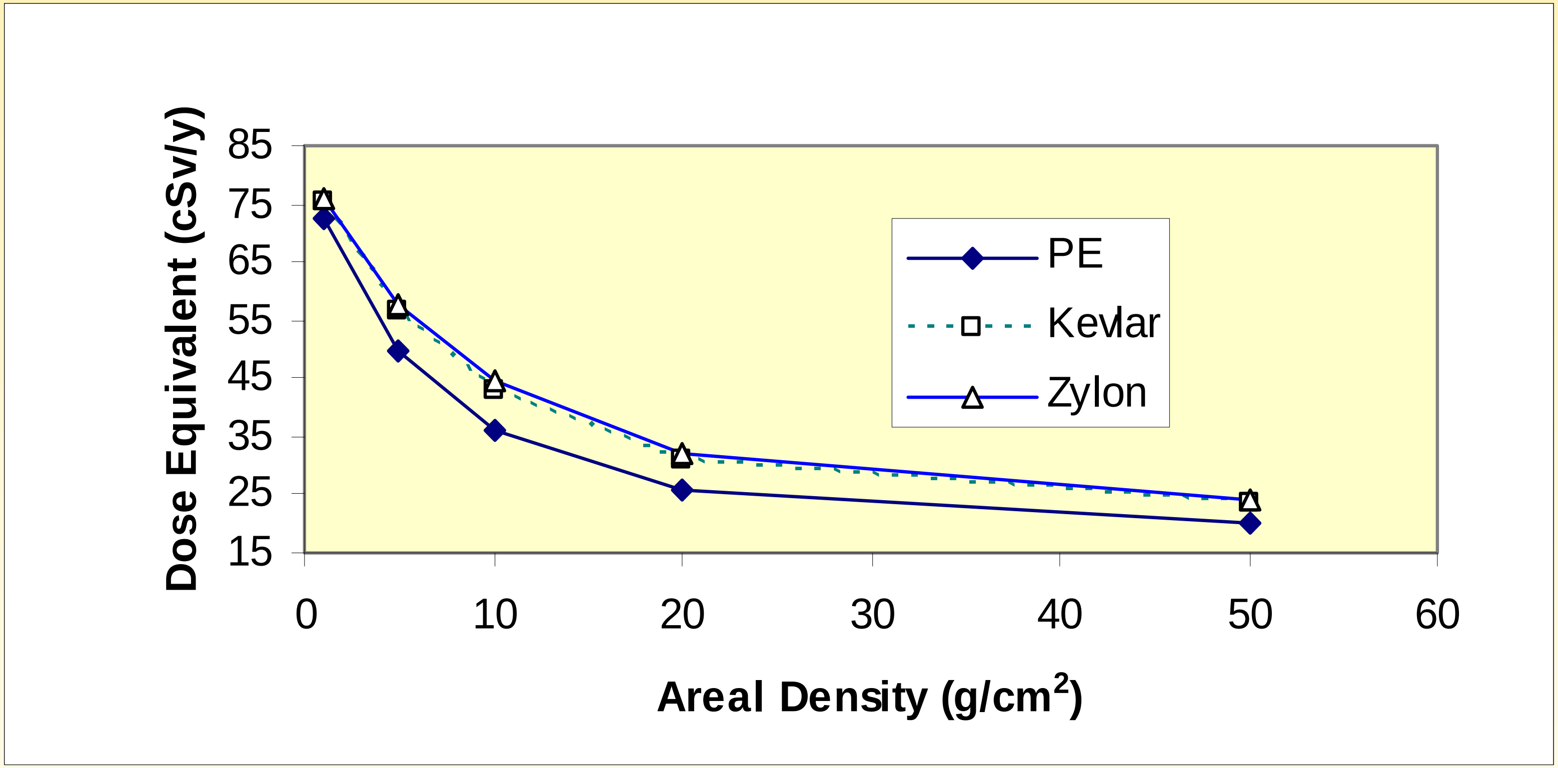
Material/Percent	Hydrogen	Carbon	Oxygen	Nitrogen	Comments
Lo Dens. Foam	10.5	89.5			
Hi Dens. Foam	7.5	92.5			
Lo Dens. Mono.	9.2	90.8			
Kevlar Fabric	4.2	70.6	13.5	11.8	
Spectra Fabric	14.3	85.7			
Zylon Fabric	2.6	71.8	13.7	12	
Spectra Epoxy	13.2	83.9	2.2	0.7	Estimated
Spectra Shield	13.0	87.0			Estimated
Zylon/PVB Phenolic	3.0	72.5	14.1	10.4	Estimated
IM 7/Epoxy	2.4	90.2	6.7		Rem. Sb, F, I, S
Epoxy	8.7	67.0	22.3	2.0	
LDPE	14.3	85.7			

Calculated Results for PE-Infiltrated Carbon Fiber Monolith



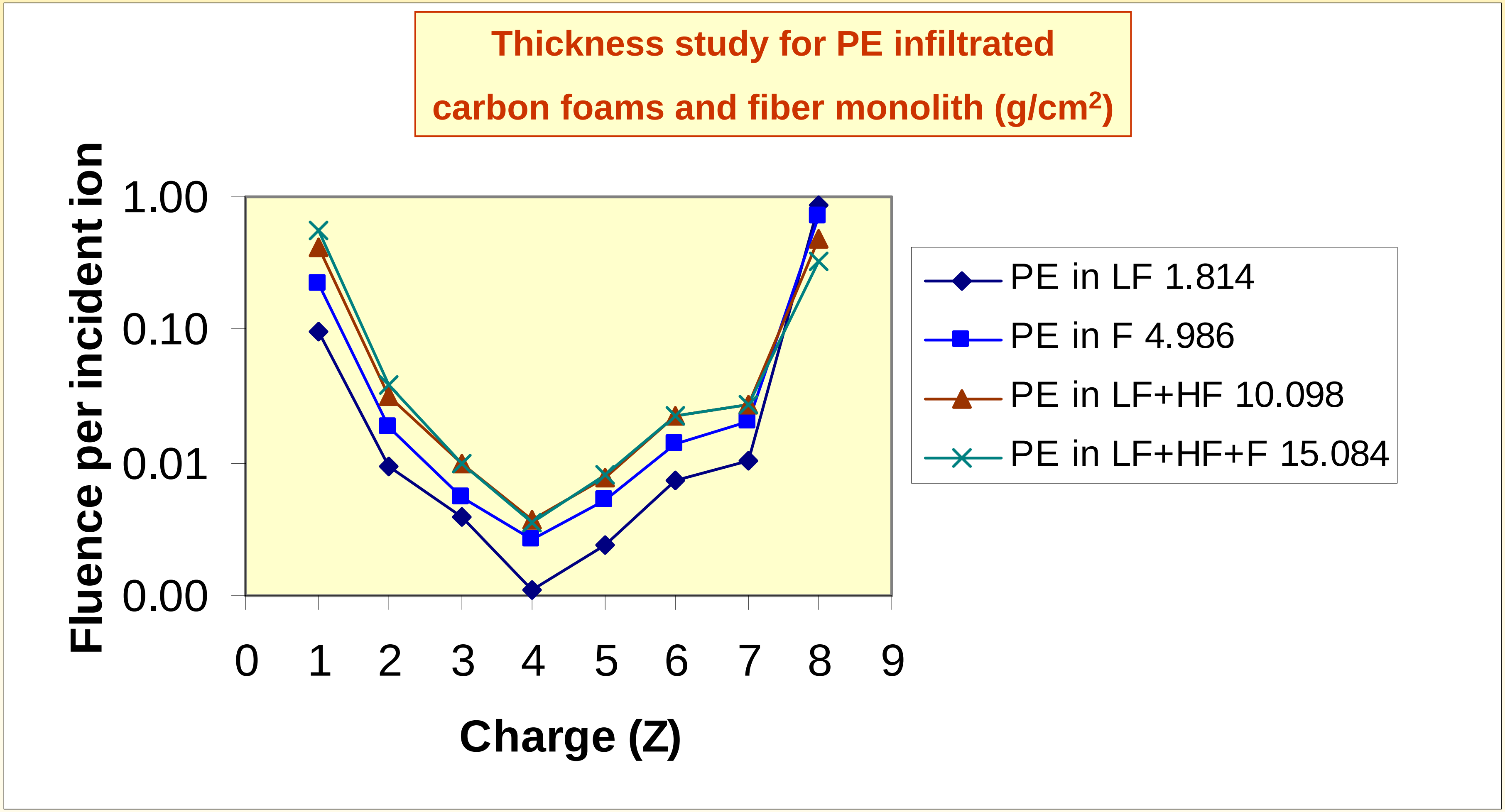
Dose equivalent to skin for the 1977 Solar Minimum GCR Spectrum. (For blood forming organs, dose equivalent ranges from ~ 43 to ~ 21 cSv/y on the above ordinate for both materials.)

Calculated Results for Polymers

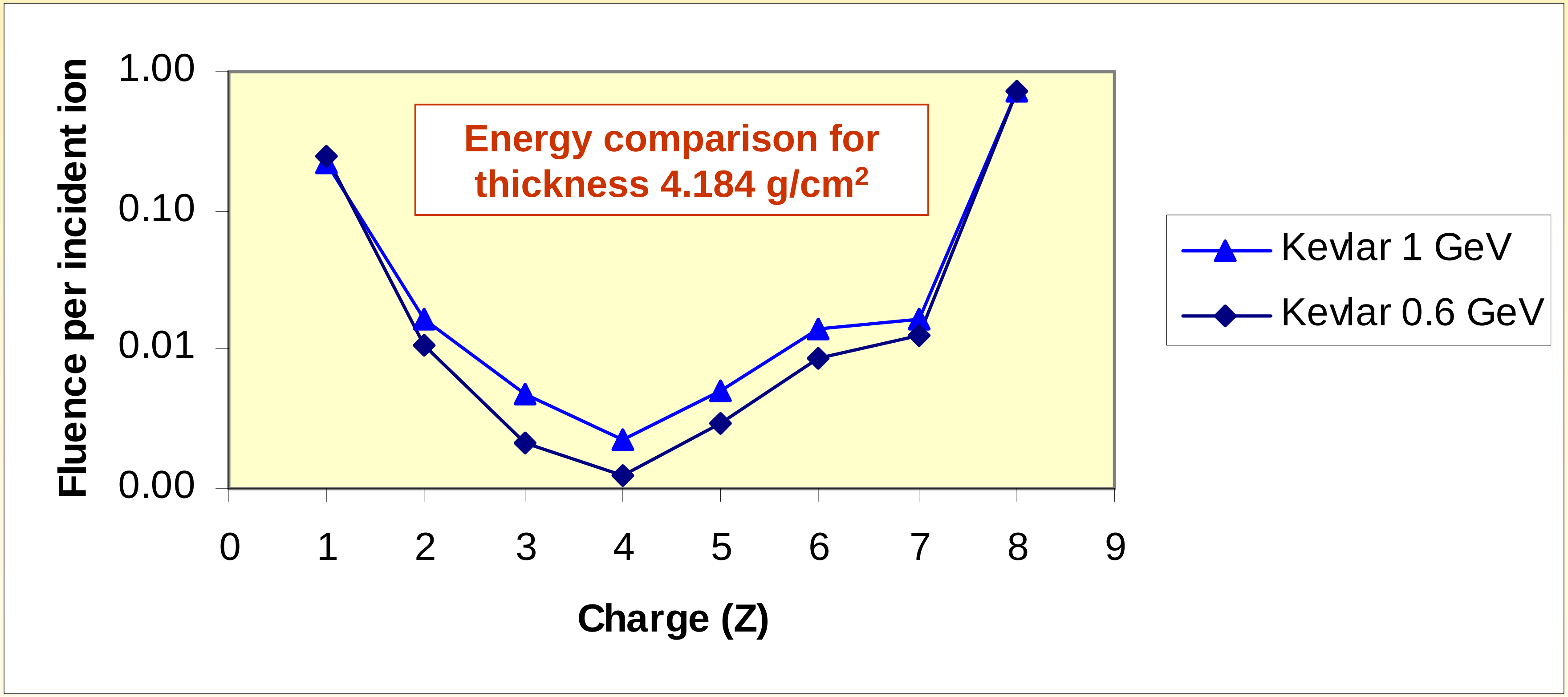


Dose equivalent to skin for the 1977 Solar Minimum GCR Spectrum. (For blood forming organs dose equivalent ranges from ~ 44 to ~ 23 on the above ordinate for both high performance polymers and from ~ 43 to ~ 21 for PE.)

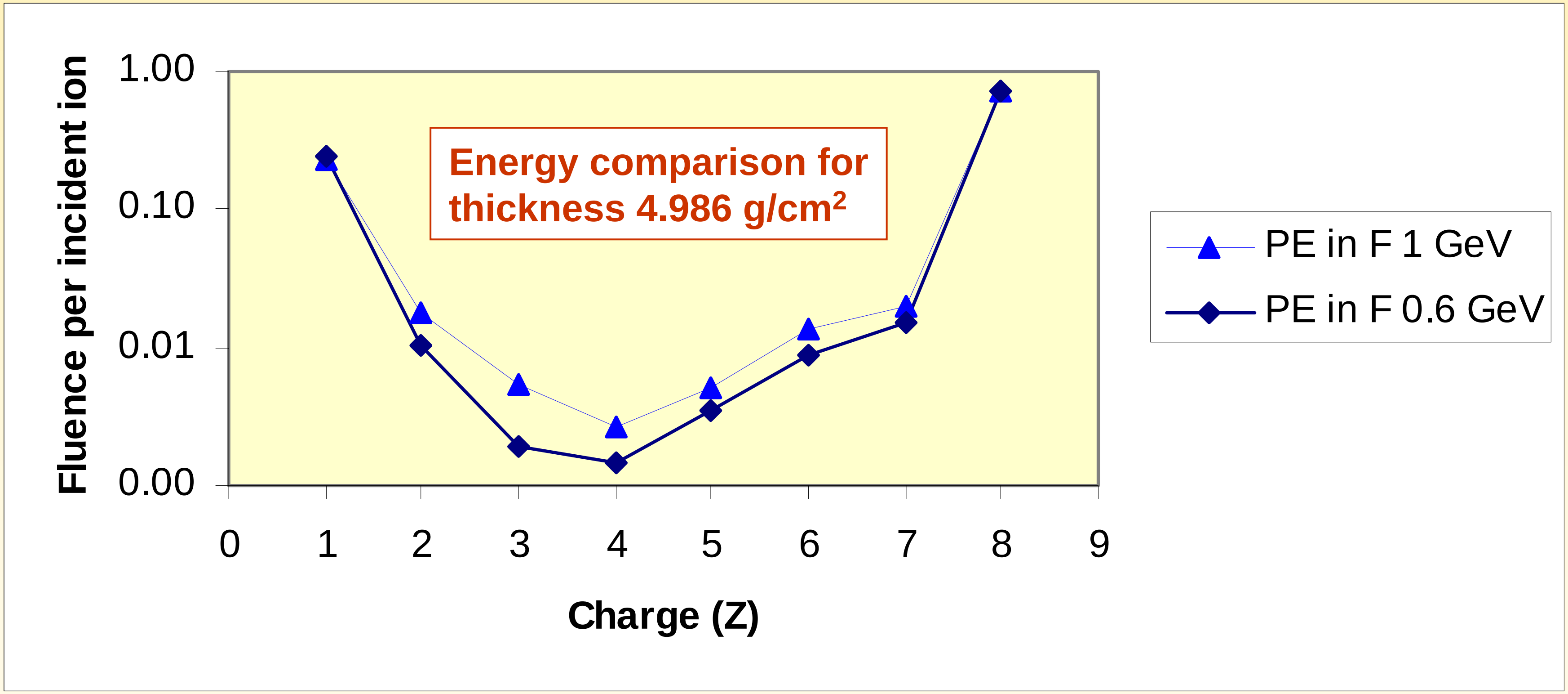
Beam Fragmentation Measurements at NSRL Using 1 GeV/nucleon O^{16}



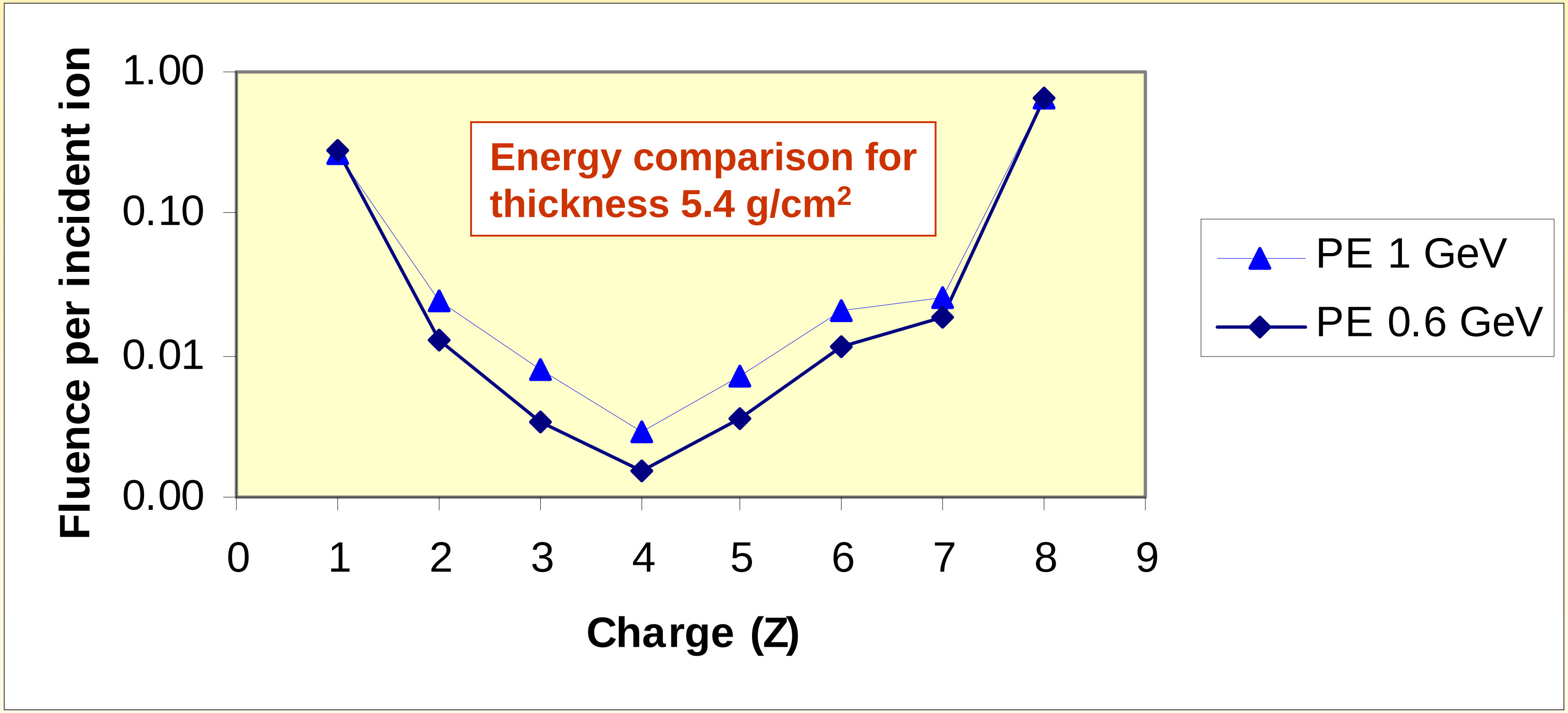
Beam Fragmentation Measurements at NSRL with 1 and 0.6 GeV/nucleon O^{16}



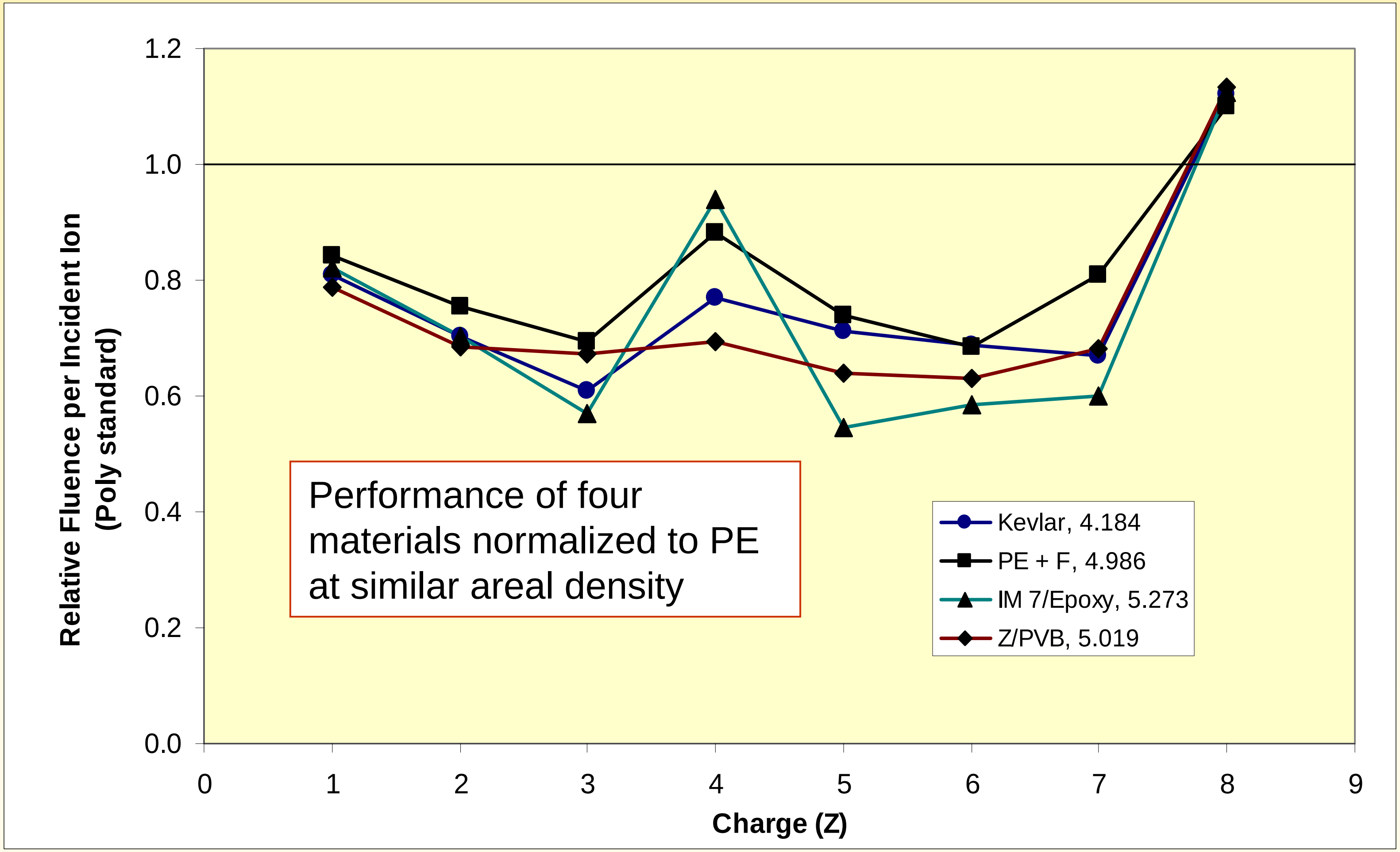
Beam Fragmentation Measurements at NSRL with 1 and 0.6 GeV/nucleon O^{16}



Beam Fragmentation Measurements at NSRL with 1 and 0.6 GeV/nucleon O^{16}



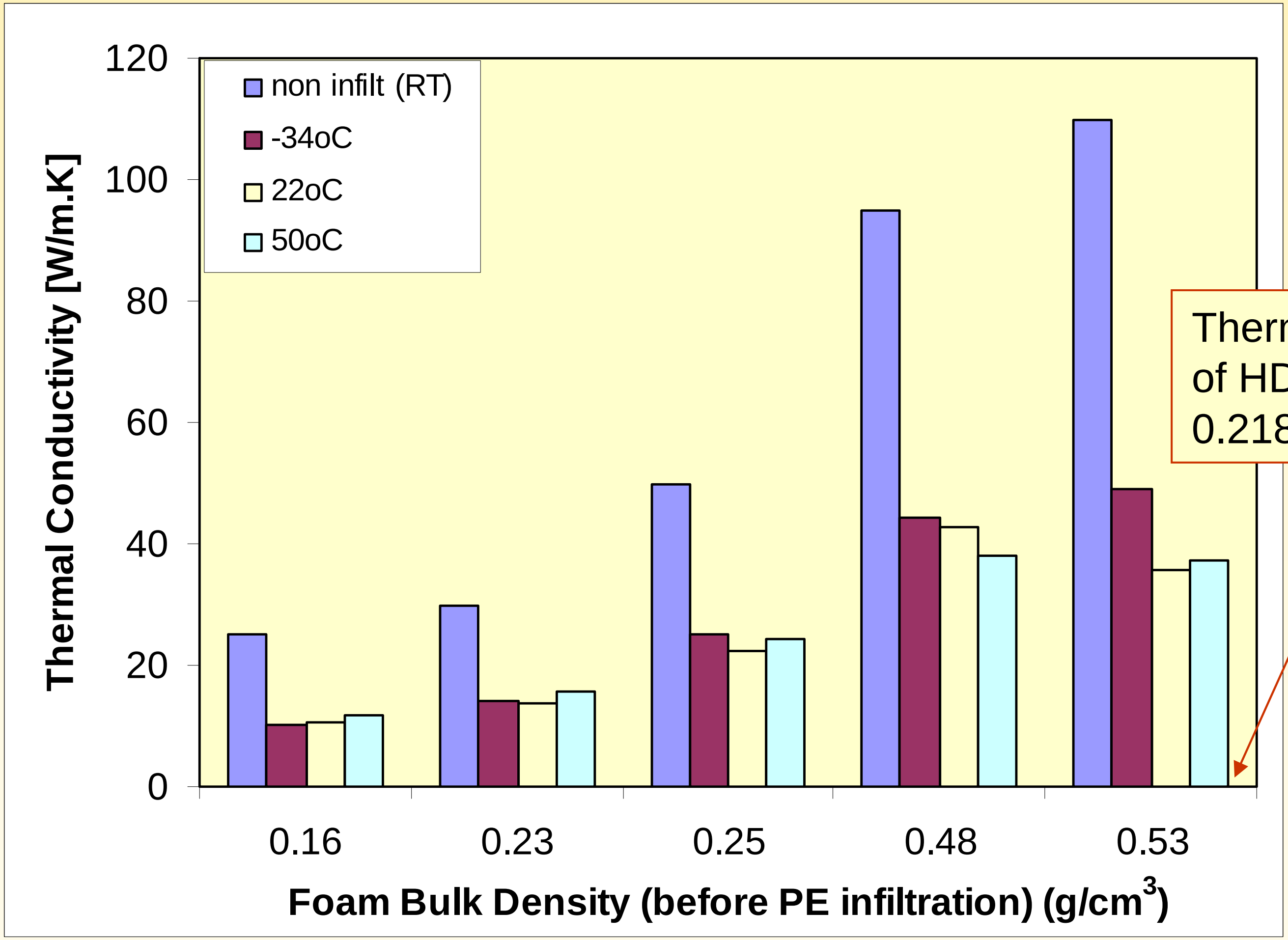
Beam Fragmentation Measurements at NSRL Using 1 GeV/nucleon O^{16}



One of the Goals--Multiple Functions, e.g., Provide Shielding and Thermal Management

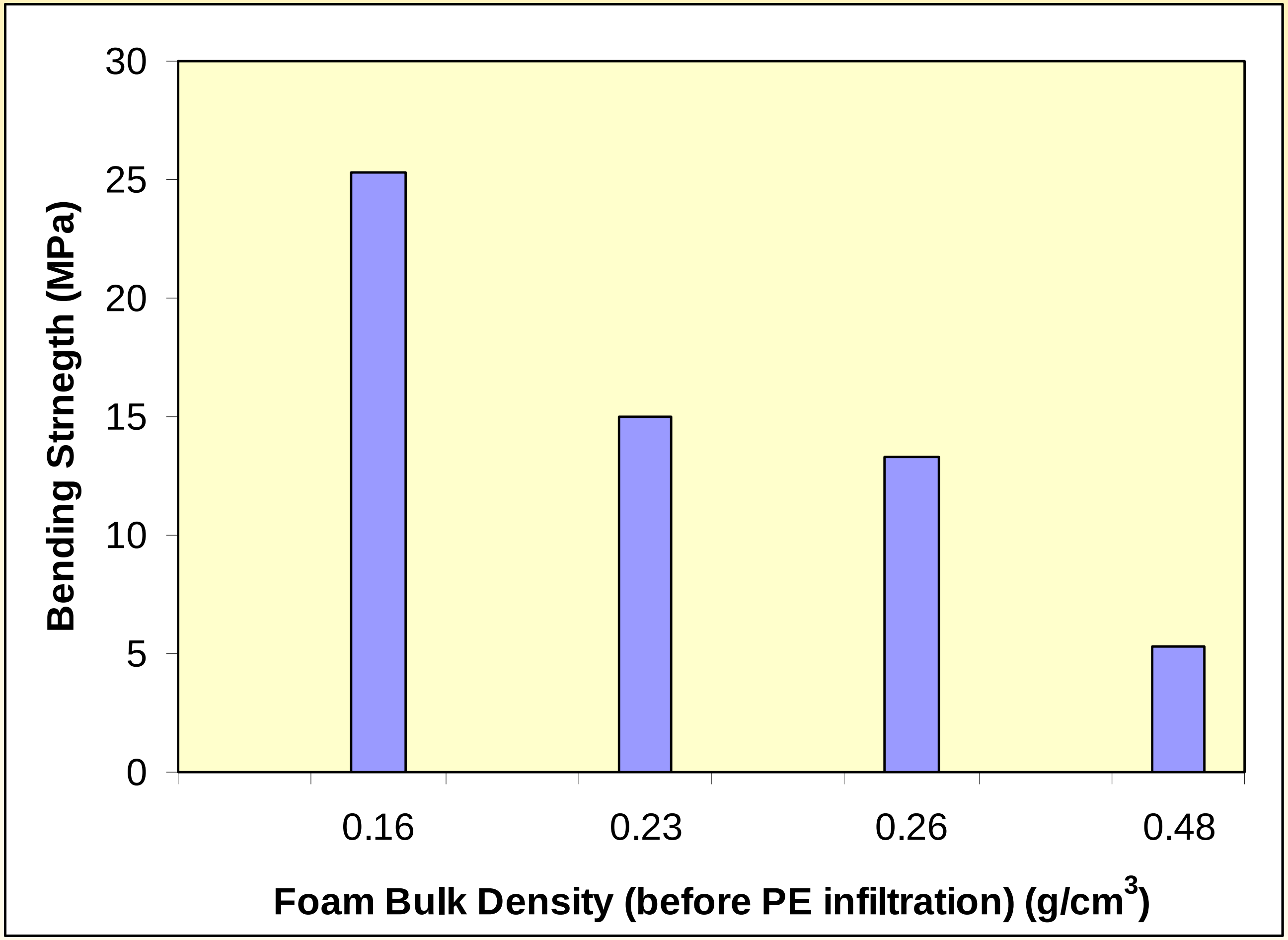
- Carbon (graphitic) foams possess high thermal conductivity
- Our NSRL work shows that PE infiltrated foams have shielding performance approaching that of PE
- Do the carbon foams retain good thermal conductivity after PE infiltration?
- What are their mechanical properties?

Thermal Conductivity Measurements of Non-Infiltrated and HDPE-infiltrated Carbon Foam

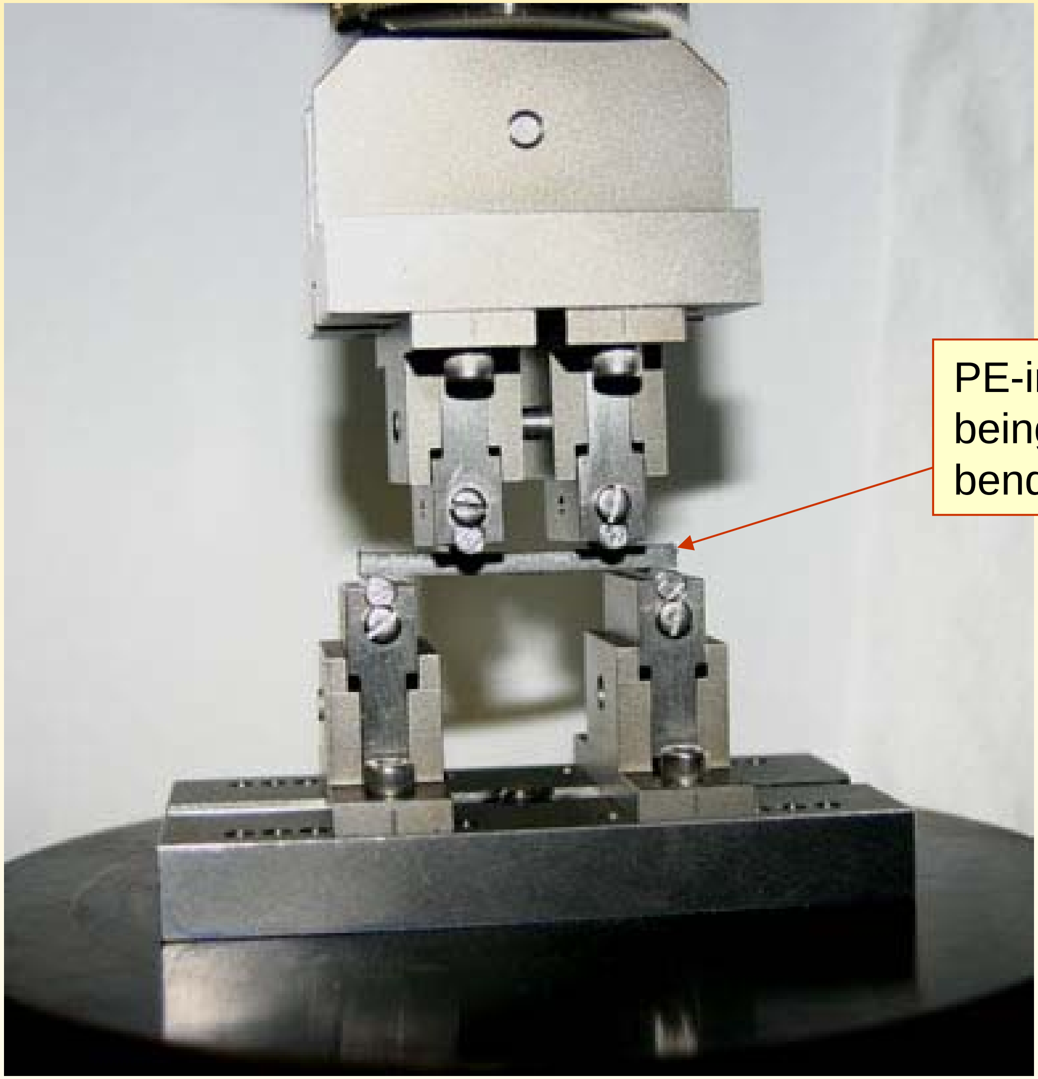


Thermal Conductivity of HDPE Infiltrate 0.218 W/m·K

Flexure Tests Results



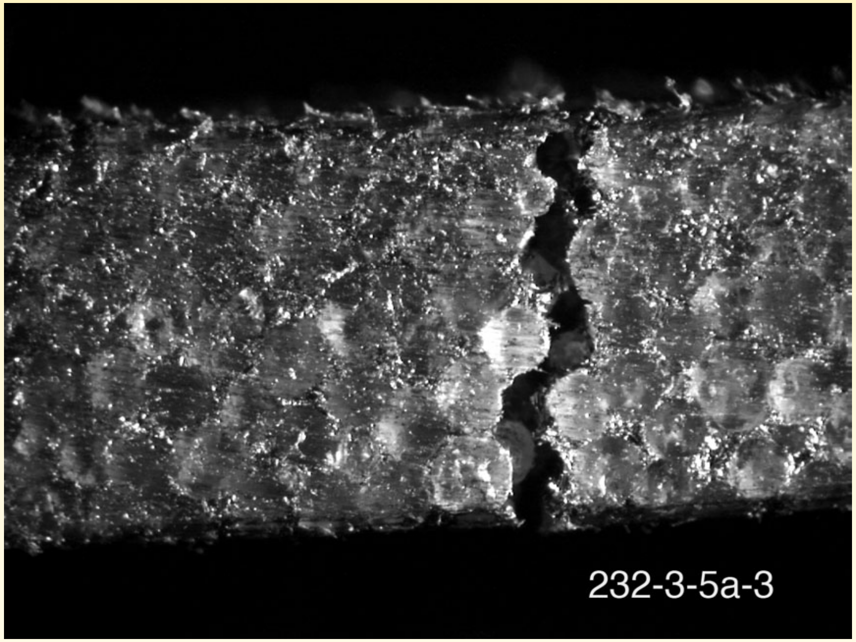
Four Point Bend Test

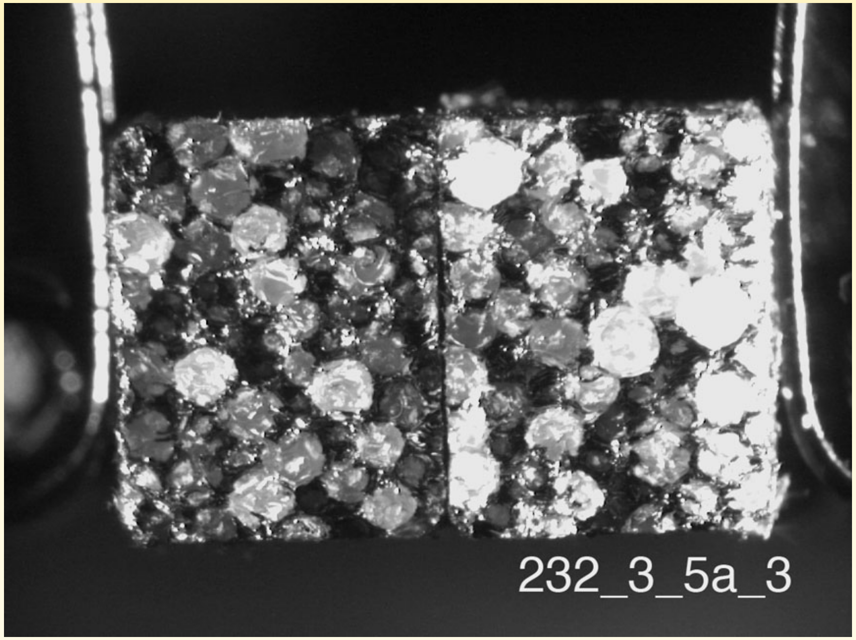


PE-infiltrated carbon foam sp
being tested in self-aligning 4
bend fixture with a 20/40mm s

Fracture Appearance after Flexure Test

Optical micrographs of failed specimen
Illustrating crack path (left) and showing
tensile sides (in center) of two halves (right).





Summary

- Twelve materials containing low atomic mass elements were prepared for measurements at NSRL
- Three were carbon forms infiltrated with PE
- Others were polymers and polymer matrix composites
- Irradiations were carried out using beams of 1 and 0.6 GeV/nucleon O^{16}
- Analyses of results show that low density carbon foam and carbon fiber monolith infiltrated with PE are next most effective in shielding to PE control
- Carbon foams retained up to half the initial high thermal conductivity after PE infiltration--possible dual use for GCR shielding and thermal management
- Preliminary mechanical property tests show that these materials could support low stresses--possible dual use for GCR shielding and low load structural applications

Radiation Effects

- Expect no significant effect of GCR on polymer properties

“Tutorial” on Radiation Effects in Materials

- **Short answer**
  - Virtually every property can be changed by irradiation
- **Long answer**
  - Dimensions
  - Mechanical properties
  - Physical properties (electrical, optical, thermal…)
  - • • •
- **Underlying these changes are the production of defects and defect clusters, alterations in microstructure (e.g., dislocations, voids, precipitates) compositional segregation, electronic ionization and excitation (breaking of chemical bonds)…**

Radiation Effects in Materials

- **Radiation damage**
  - Key issue is usually cumulative degradation, although instantaneous effects can be important in some cases (example: radiation induced conductivity in insulators)

Historical Perspective on Radiation Effects in Materials

- **Some radiation effects were observed in minerals in the 19th century, but their origin was not understood**
- **E. P. Wigner, 1946, Journal of Applied Physics 17**

  "The matter has great scientific interest because pile irradiation should permit the artificial formation of displacements in definite numbers and a study of the effect of these on thermal and electrical conductivity, tensile strength, ductility, etc. as demanded by the theory."

- **The full scope of radiation effects in materials was only appreciated after high neutron flux fast spectrum reactors were operated in the 1950's and 1960's**
- **Targets of present and planned high beam intensity accelerators are roughly at the same levels of damage rate as the highest flux fission reactor cores**

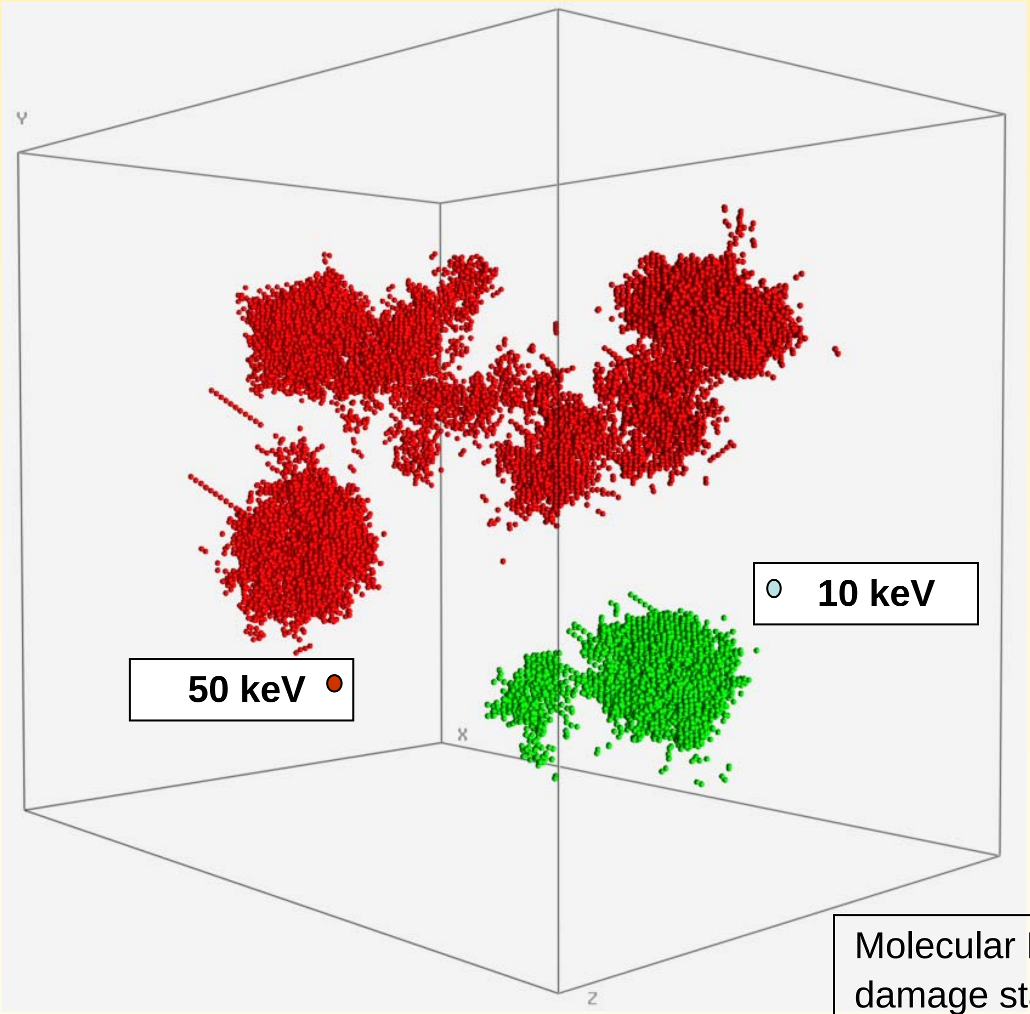
Origins of Radiation Effects in Materials

- **Displacement of atoms (nuclear stopping)**
  - Dominant damage process for metals
  - Important for ceramics, semiconductors
  - Could be significant for polymers (usually neglected)
  - Dose unit--displacement per atom, *dpa*
  - One dpa is the dose at which on average every atom in the material has been energetically displaced once
- **Ionization and excitation (electronic stopping)**
  - Generally can be neglected for metals
  - Important for polymers
  - Can be important for ceramics, semiconductors
  - Dose unit--Gray, *Gy*, the dose for absorption of 1 J/Kg

Origins of Radiation Effects in Materials

- **Transmutation reactions**
  - Transmutation products, especially helium and hydrogen from proton- and neutron-induced reactions, exacerbate damage in metals and ceramics
  - Customary unit of measure is appm transmutant per dpa, e.g., *appm He/dpa*
- **Typical highest average damage rates (10^{-6} dpa/s, $>10^{3}$ Gy/s, 100 appm He/dpa)**
  - High flux reactor core
  - Fusion reactor first wall
  - High power spallation target

Displacement Damage Occurs in Cascades



- High energy particles, e.g., GeV particles, spallation neutrons, or d-t fusion neutrons produce atomic recoils at much higher energies than fission neutrons
- Large-scale atomic simulations demonstrate that subcascade formation leads to similar defect production
- Subcascades from 50 keV event (avg. from 2.3 MeV neutron) are similar to a single 10 keV event (avg. from 0.4 MeV neutron)
- Average defect production per unit cascade energy is essentially the same for recoil energies above tens of keV

Molecular Dynamics Simulations of peak damage state in iron cascades at 100 K
R. E. Stoller

Time and Energy Scales for Radiation Effects by Displacement Damage

Time	Energy
Cascade Creation 10^{-13} s	Neutron or Proton 10^{6} - 10^{9} eV
Unstable Matrix 10^{-11} s	Primary Knock-on 10^{4} - 10^{5} eV
Interstitial Diffusion 10^{-6} s	Displaced Secondary 10^{2} - 10^{3} eV
Vacancy Diffusion 10^{0} s	Unstable Matrix 10^{0} eV
Microstructural Evolution 10^{6} s	Thermal Diffusion kT

Basics of Radiation Effects on Polymers

- **Comparatively low doses can produce changes in properties**
  - Why? Because of typically very high molecular weight, a large fraction (tens of percent) of the molecules can suffer at least one ionization event in doses of order 10 kGy
- **Predominant changes can be described as chain scission and cross-linking (other changes: release of small molecules, i.e., gas formation, modification in types of bonding, …)**
- **For a given polymer, radiation type and temperature, either cross-linking or scission will usually dominate**
  - Cross-linking increases molecular mass, lowers solubility and improves mechanical properties
  - Scission generally degrades properties

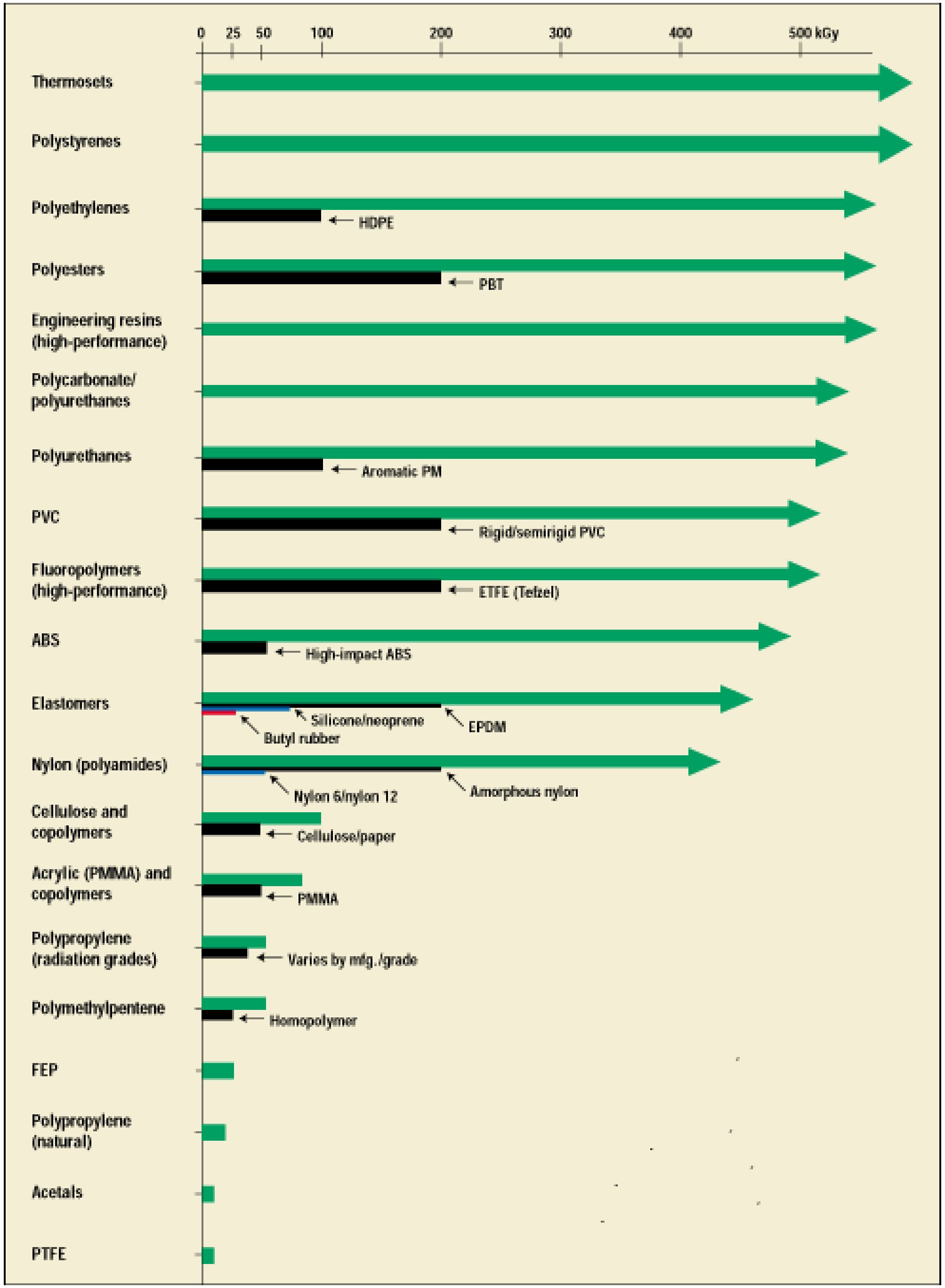
Basics of Radiation Effects on Polymers

- **Polymers have been broadly classed in the literature as cross-linking type or degrading (scission) type**
- **Our research showed that the ratio of cross-links to scissions depends strongly on linear energy transfer (LET). Energetic heavy ions cause much more cross-linking than γ or e^- and can lead to reclassification of a material from scission- to cross-linking type**
- **Range of sensitivity for producing significant degradation spans many orders of magnitude in dose, for example, for reduction in uniform elongation in a tensile test**

  $<$ 1 kGy PTFE (Teflon)

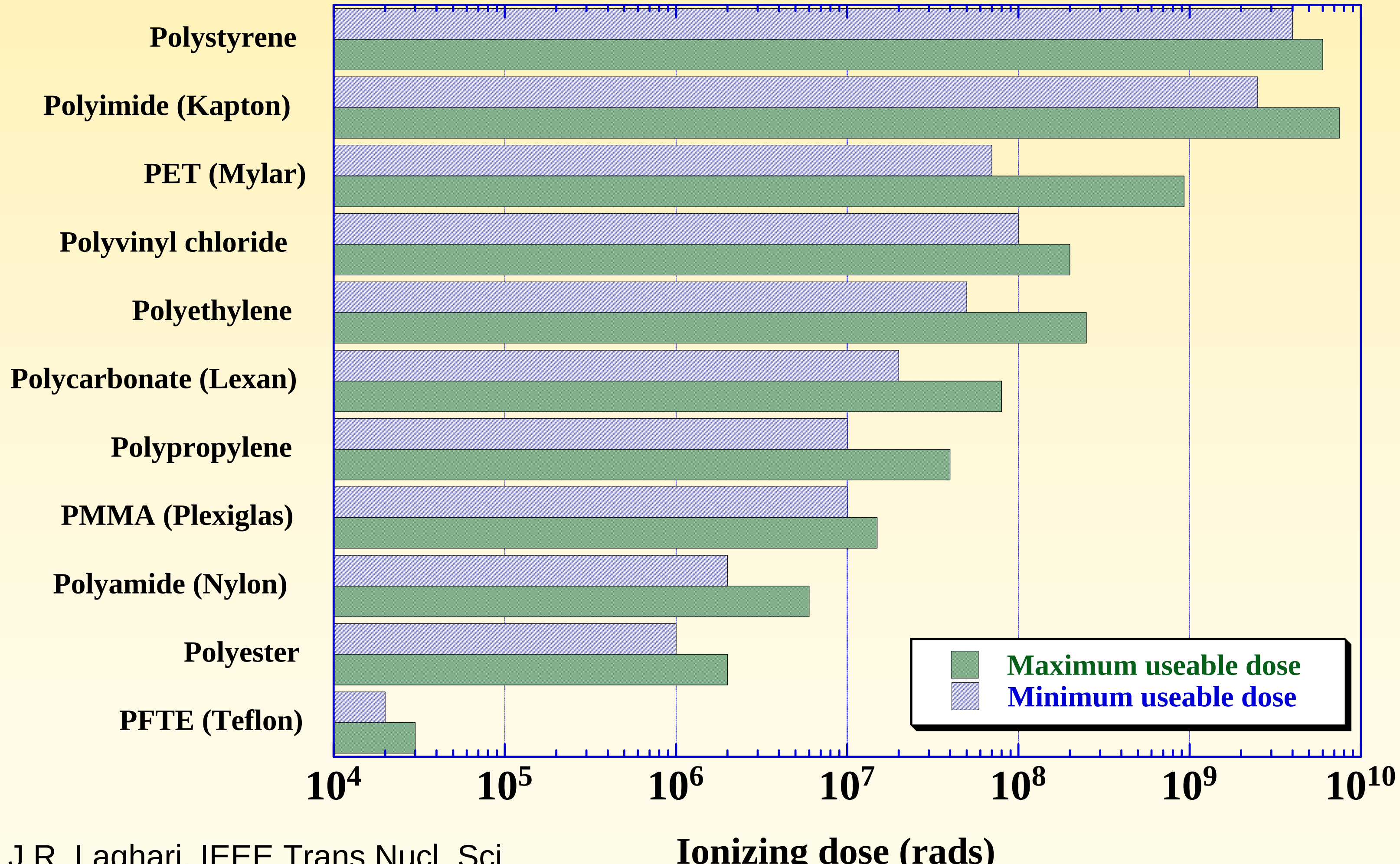
  $\gtrsim 10^3$ kGy PI, PS (Polyimide, Polystyrene)
- **Sensitivity also depends on irradiation conditions and environment. Irradiation in vacuum can improve dose endurance over that in air by an order of magnitude. Irradiation at higher T also can give improvement.**

Mechanical Properties of Polymers (dose to reduce elongation by 25%)



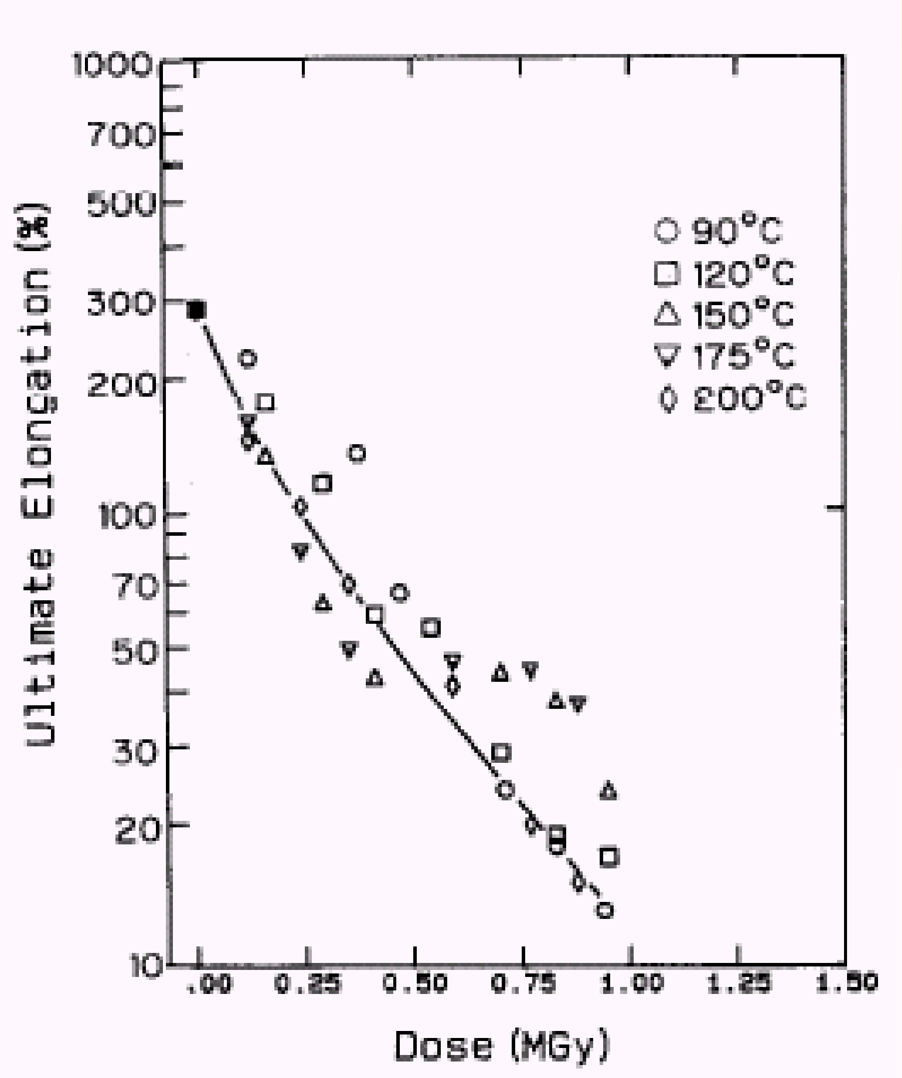
K. J. Hemmerich, Med. Dev.
& Diag. Ind. Magazine, Feb. 2000

Summary of Radiation Dose Limits for Polymers



J.R. Laghari, IEEE Trans Nucl. Sci. 37, 2 (1990)1076

Decrease in Elongation of Viton Elastomer Irradiated at Various Temperatures



M. Ito, Radiat. Phys. Chem. 47(1996)607-610

Conclusions

- **Radiation effects in polymers become significant over the range from < 1 kGy to > 1 MGy, depending on the material**
  - Even for minimal radiation fields, acetals, polypropylene, and PTFE (Teflon) should be avoided
  - Top performers are PI (polyimide) and PS (polystyrene)
  - High performance fluoropolymers like Viton are in an intermediate range. However, “Viton” is a general name for entirely different formulations. Specific data for the precise formulation should be consulted.

Approximate Radiation Dose Limits

- **People < 1 Gy (Sv) (ALARA)**
- **Polymers: 10^2 to 10^7 Gy**
- **Semiconductors: ~10^{13} n/cm^2, ~10^2 Gy (10^{16} to 10^{17} for SIC JFETs at 300˚C)**
- **Piezoelectric crystals: 10^{14} to 10^{19} (?) n/cm^2**
- **Ta capacitors: ~ 10^{15} n/cm^2, ~10^5 Gy**
- **Organic lubricants: 10^{16} n/cm^2, ~10^6 Gy**
- **Graphite, $MoSi_2$ lubricants: ~ no degradation up to 10^{19} n/cm^2**
- **Magnets: 10^{18} n/cm^2: up to 30% increase in coercive force and magnetic remanence**
- **Glass: 10^{20} n/cm^2 (>10% dimension change); 10^8 Gy (optical darkening saturates)**
- **Ceramics:**
  - ~10^9 Gy, ~ 10^{20} n/cm^2 (radiolysis-sensitive ceramics)
  - >10^{21} n/cm^2 (> 1 dpa) for most oxides, carbides and nitrides
- **Metals: > or >> 10^{21} n/cm^2 (>1 dpa); ionizing radiation ~ negligible**

Thanks to S. J. Zinkle, ORNL

Rough Dose Equivalences for Materials

- Fast fission reactor spectrum: $1x10^{10}$ n/cm^2 ~2 rads (centiGray) ~ $0.8x10^{-11}$ dpa (equal contributions from gamma ray and neutron pka ionization)
- Mixed spectrum reactor: $1x10^{10}$ n/cm^2 ~ 40 rads (centiGray) ~ $0.8x10^{-11}$ dpa (ionization dose mainly due to gamma rays, precise values depend on reactor design and material)

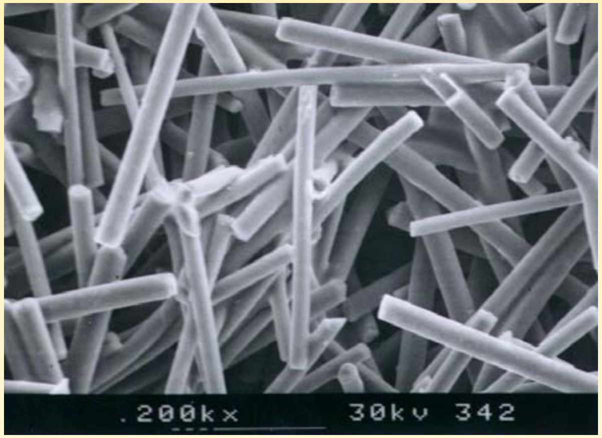
Methane Adsorbed on Carbon Forms as a Possible Alternative for GCR Shielding

- Advantage of finely divided carbon for methane storage--pressure reduced by an order of magnitude.
- Previously we evaluated fiber monoliths for room-T methane storage. Best monolith had storage of 13.16 wt %, at 3.4 MPa (~ 33 bar).
- Adsorption capacity increases as temperature decreases, but no quantitative data were available.
- Status--New experiments have been completed to measure gravimetric methane storage capacity of carbon materials at lower temperatures.

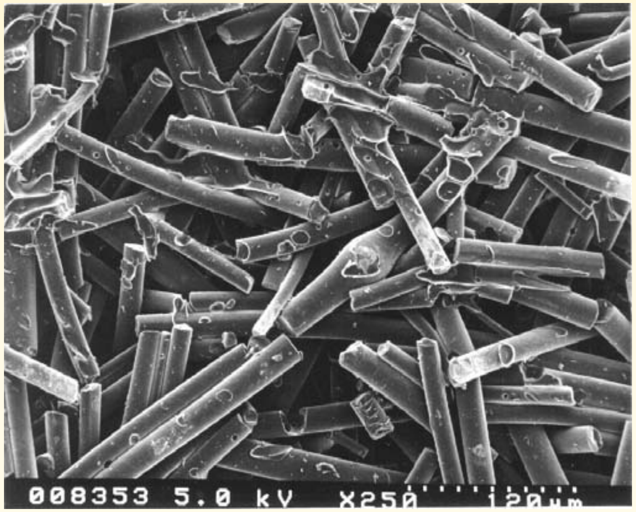
Specimens Tested

Material	Density [g/cc]	Area [m^2/g]
Fiber Monolith SMS-22	0.37	2451
Fiber Monolith SMS-50	0.56	2020
Powdered Carbon – Westvaco	0.30	2200

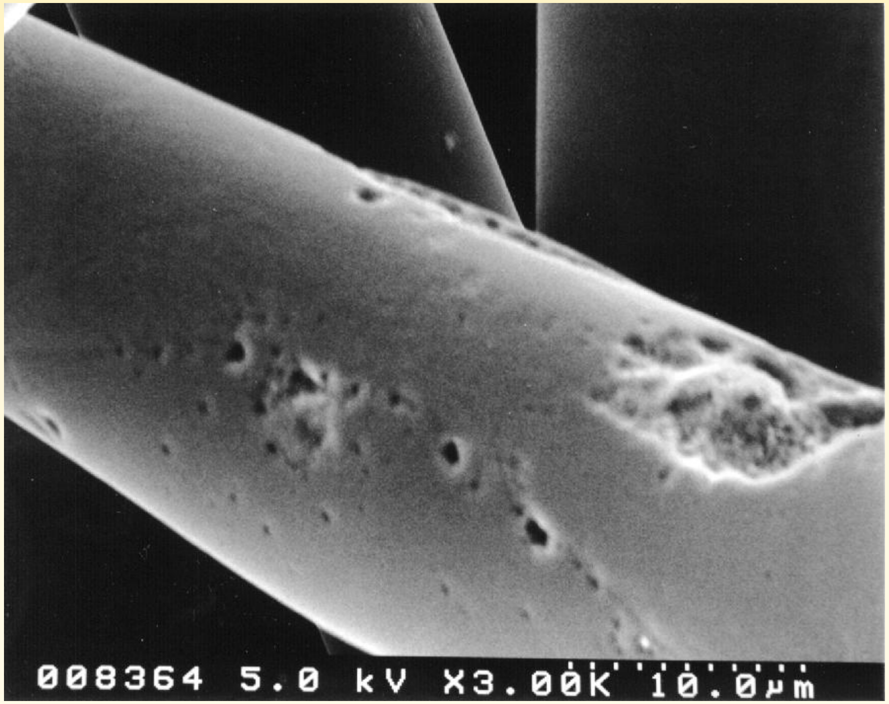
Structure of Fiber Monoliths

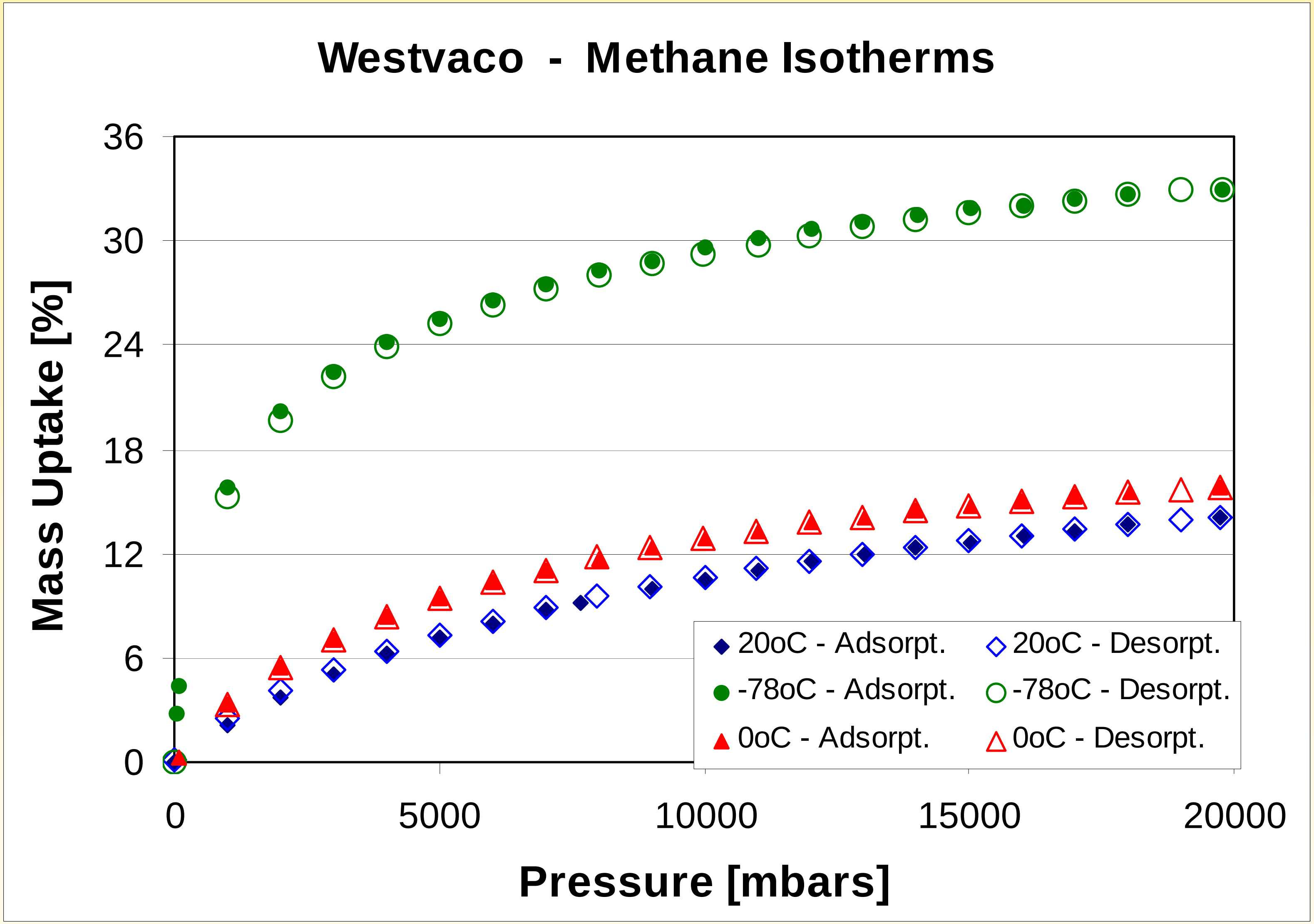


Low Density Monolith



High Density Monolith

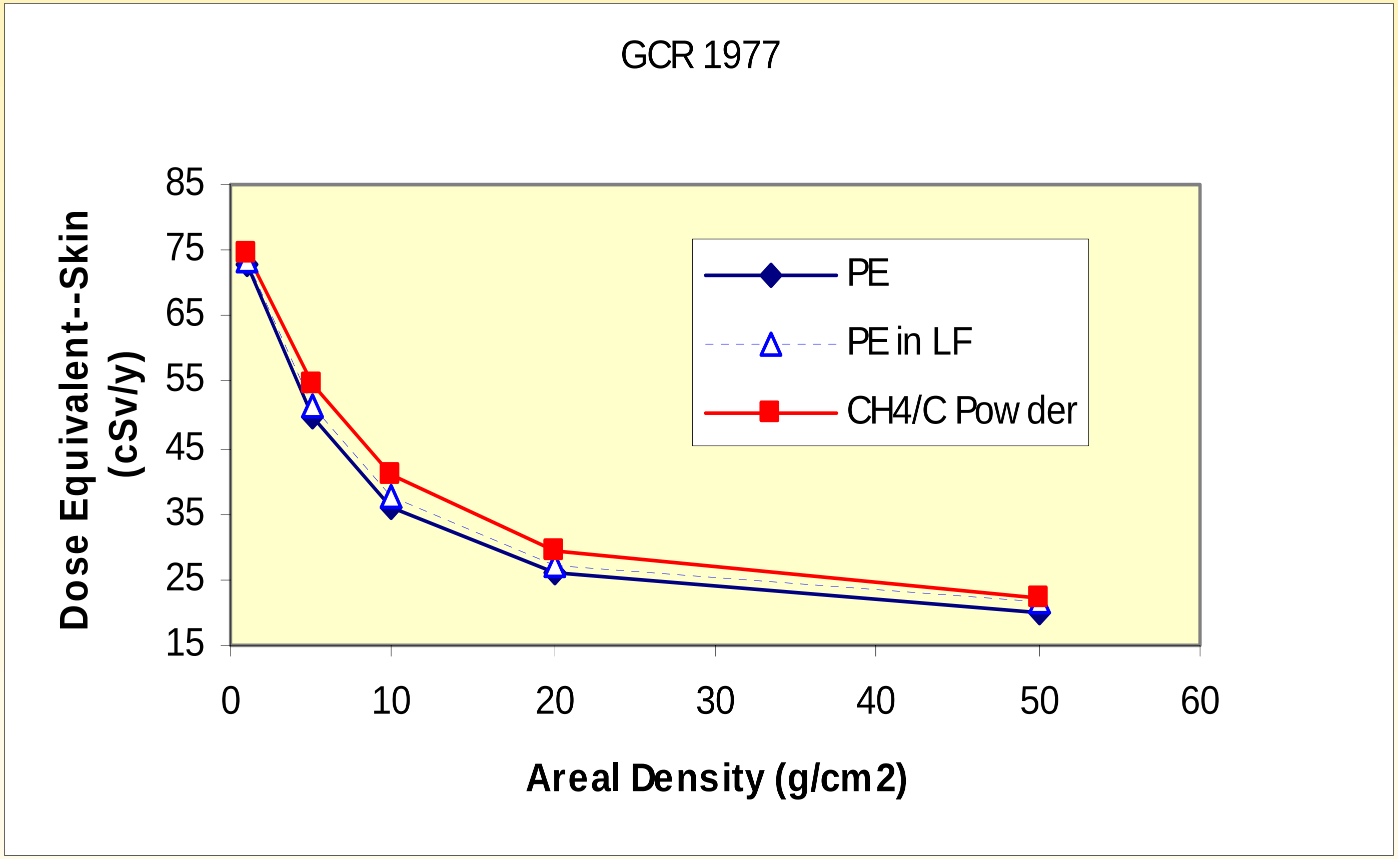



Westvaco - Methane Isotherms
Mass Uptake [%]
Pressure [mbars]
0
6
12
18
24
30
36
0
5000
10000
15000
20000
20oC - Adsorpt.
20oC - Desorpt.
-78oC - Adsorpt.
-78oC - Desorpt.
0oC - Adsorpt.
0oC - Desorpt.

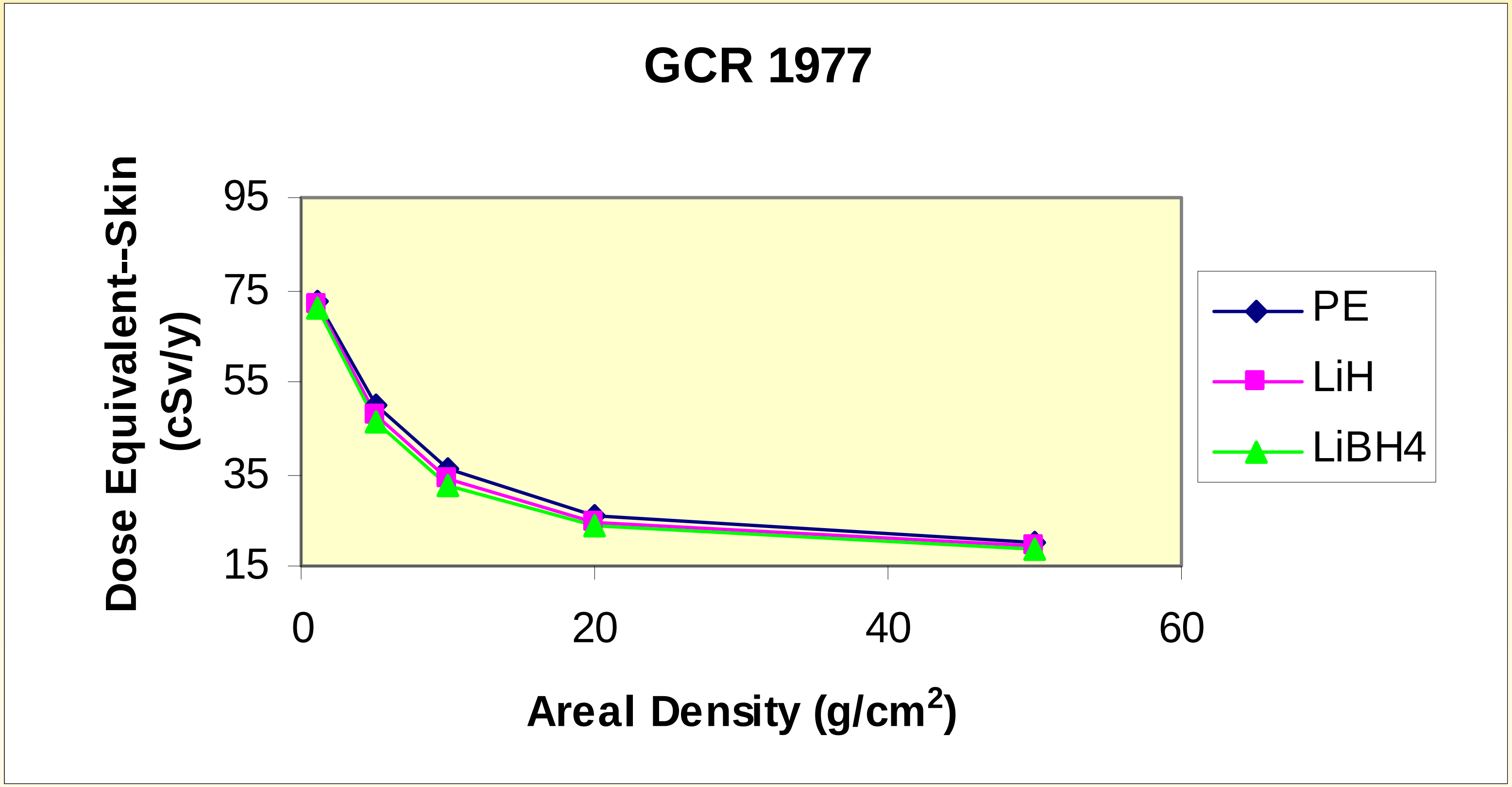
Summary of T- dependence Experiments on CH_4 Adsorption in Carbon Materials

- **Significant improvement in methane adsorption is observed by reducing the temperature to ~ -78^{o}C.**
- **The Westvaco material exhibited the greatest methane uptake at 20^{o}C and -78^{o}C.**
- **The maximum uptake of methane at -78^{o}C and <u>20 bars</u> was <u>6.2 wt% hydrogen.</u>**

GCR Shielding Performance for PE-infiltrated Carbon Foam and Best CH_4-Adsorbing Carbon



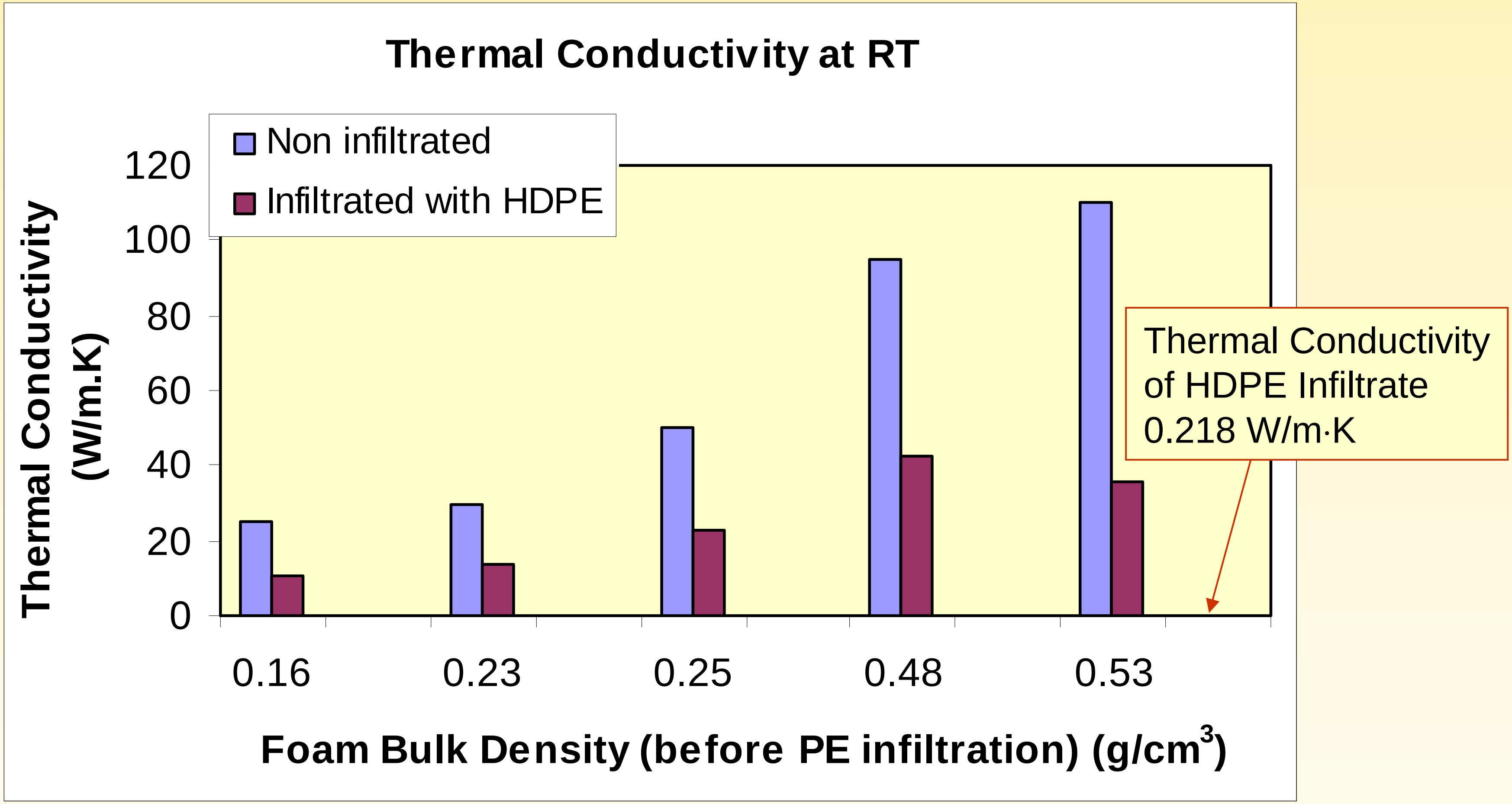
GCR Shielding Performance for Lithium Hydrides



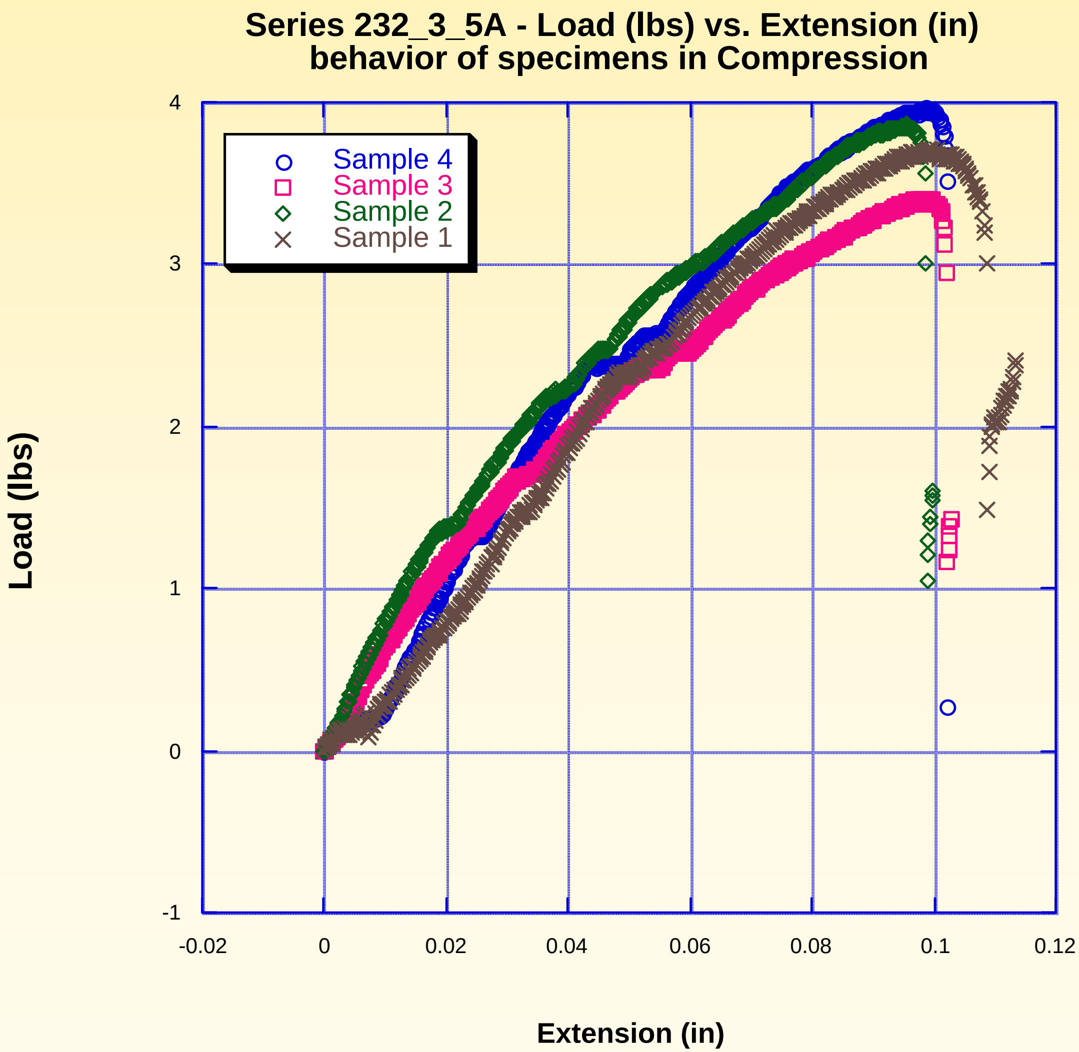
Polymer Fibers and Composites

- **Spectra: Polyethylene CH_2**
- **Kevlar: Poly(p-phenylene terephthalamide) $C_{14}H_{10}O_2N_2$**
- **Zylon: Poly(p-phenylenebenzobisoxazole) $C_{14}H_6O_2N_2$**
- **M5: Poly{2,6-diimidazo[4,5-b4',5'-e]pyridinylene-1,4(2,5-dihydroxy)phenylene} $C_{13}H_7O_2N_5$**

Carbon Foams Retained up to Half Their Initial Thermal Conductivity after PE Infiltration



Shape of Compression Test Curves



Sample 232-3-5A

1. Foam bulk density before PE infiltration: 0.23 g/cc
2. % void fraction: 89%